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Phonon dispersion and electrical resistivity of sodium using an angular force model

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A five parameter model for bcc metals is proposed by combining ion-ion central, ion-ion angular and electron-ion interactions. The electron-ion interaction which has been included on the lines of Sharma and Joshi's theory, employs a correct expression for the G-function of Bardeen. The phonon dispersion curves and the temperature variation of the electrical resistivity of sodium have been investigated using the modified model. The theoretical results are in reasonable agreement with the experimental data.

1. INTRODUCTION

The main complexity in the rigorous treatment of the lattice dynamics of metals is concerned with the effect of conduction electrons on the lattice vibrations which is basically a many body problem. Nardeen (1937) while establishing the validity of Bloch-Wilson theory of the electrical conductivity of metals, dealt with the theory of electron-ion interaction in metals. Toya (1958) extended the Bardeen's theory to include the correlation and the exchange energies of valence electrons. However, Toya's theory could not predict the quantitative features of the experimental specific heat versus temperature curve which was attributed to the use of Bardeen's G-function in the expression for electron-ion interaction by Dayal & Shrivastava (1964). The expression for the G-function has been derived within the free electron approximation by Bardeen (1937) on an assumption that the atomic polyhedron can be replaced by a sphere of equal volume.

Recently Bross & Bhon (1967) have derived an exact expression for Bardeen's G-function by considering the actual size of Wigner-Seitz cell for fcc and bcc structures. They retain all the essential features of the original Bardeen matrix elements except that wherever the Wigner-Seitz sphere enters into picture (in the definition of the potential, in the integration etc.) it is replaced by the Wigner-Seitz polyhedron. Therefore, it is imperative to use the correct expression for the G-function in the study of lattice dynamics of metals (Goel *et al* 1974).

The purpose of the present paper is to propose a lattice dynamical model for bcc metals and to study the phonon dispersion relations and the electrical resistivity of sodium as an application of the proposed algorithm. The model

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employs the central forces and the angular forces (Clark *et al* 1964), both up to second neighbours and the electron-ion interaction has been included on the lines of Sharma & Joshi's (1963) model with an exact expression for Bardeen's G-function. The computed phonon dispersion relations and the electrical resistivity of sodium are found to be in good agreement with the experimental data.

2. Theory

The normal modes of vibrations ω are obtained by solving the secular equation

$$|D(q)-m\omega^2 I| = 0, \qquad \ldots (1)$$

where m is the mass of the atom, I is a 3×3 unit matrix and $D(q)$ is the dynamical matrix. The elements $D_{ij}(q)$ $(i, j = 1, 2, 3)$ of the dynamical matrix $D(q)$ can be taken as sum of three terms arising from the ion-ion central interaction, the ion-ion angular interaction and the electron-ion-interaction. The typical diagonal and non-diagonal elements of the dynamical matrix of a bcc lattice can be written as

$$D_{ii}(q) = 8(A_1+2K_1+3K_2)(1-C_iC_jC_k)+4(A_2+4K_1)S_i^2+ \\ +2(3K_2-2K_1)(S_2^2+S_3^2)+a\pi^2K_eG^2(q)q_i^2 \qquad \ldots (2)$$

$$D_{ij}(q) = 8(A_1-K_1+3/2K_2)S_iS_jC_k+a\pi^2K_eG^2(q)q_iq_j, \qquad \ldots (2)$$

where $C_i = \cos(\pi q_i)$, $S_i = \sin(\pi q_i)$, q_i are the Cartesian components of the reduced wave vector and a is the lattice parameter; A_1 and A_2 are the first and second neighbour central force constants and K_1 and K_2 are the corresponding angular force constants. K_e is the bulk modulus of the electron gas. The function $G(q)$ may be expressed as

$$G(q) = \frac{2}{(u^2-v^2)(u^2-w^2)}\left[u\sin\frac{u}{2}(\cos u+\cos v+\cos w)\right. \\ \left.+(u\sin u+v\sin v+w\sin w)\cos\frac{u}{2}-4\sin\frac{3u}{2}\right] \\ +\text{two additional cyclic permuted terms.} \qquad \ldots (4)$$

where $u = \pi q_1$, $v = \pi q_2$, and $w = \pi q_3$. The force constants A_1, A_2, K_1, K_2 and a K_e are related to the three elastic constants c_{11}, c_{12} and c_{44} and the two zone boundary transverse frequencies in (100) and (111) direction by the following relations

$$a\,c_{11} = 2A_1+2A_2+12K_1+6K_2+aK_e \\ a\,c_{12} = 2A_1-6K_1-3K_2+aK_e \\ a\,c_{44} = 2A_1+2K_1+9K_2 \\ m\omega_{T_1}^2 = 16(A_1+2K_1+3K_2)+aK_e\pi^2G^2(q) \\ m\omega_{T_2}^2 = 4(2A_1+A_2+6K_1+9K_2). \qquad \ldots (5)$$

The numerical values of the five model parameters can be computed by using the above relations.

For a lattice of cubic symmetry, the first order variational solution of the Boltzmann equation giving the electrical resistivity of monovalent metals due to phonon scattering can be written as (Kumar & Tolpadi 1975, Kumar 1975),

$$\rho = \frac{3h}{4uK_B e^2 V_F^3 k_F^3 mNT} \sum_p \int d\Omega \int \frac{K^5(1-U^2)^4(K.e_{qp})^2 C^2(K)}{(\exp(\beta\omega_{qp})-1)(1-\exp(-\beta\omega_{qp}))}\, dK \quad \ldots (6)$$

where e is the electronic charge, K_B the Boltzmann constant, k_F the Fermi radius, N the number of unit cells per unit volume, T the absolute temperature and ω_{qp} and e_{qp} are the angular frequency and the polarisation vector for phonon wave vector q and mode of vibrations p respectively; v_F is the velocity electron, $\beta = \hbar/K_B T$, $U = K/2k_F$ and K is the scattering vector. $C(K)$ is the Bardeen (1937) matrix element for the transition of electron from initial state k to final state k' and for a free electron model it is given by

$$C(K) = \frac{[V_r - E]K^2 + W(K)q_s^2}{q^2 + K^2}\, G(q), \quad \ldots (7)$$

where $[E - V_r]$ is the kinetic energy of an electron in the lowest state at the surface of atomic polyhedron and q is the screening parameter gives by

$$q_s^2 = \frac{4ne^2}{W(K)}$$

with n being the electron density and

$$W(K) = \frac{3}{2} E_F \left(\frac{1}{2} - \frac{4k_F^2 - K^2}{8k_F K} \ln \left| \frac{2k_F + K}{2k_F - K} \right| \right)^{-1}$$

The Bardeen's matrix element $C(K)$ (eq. 7) has been evaluated using a correct expression for G-function given in eq. 4.

3. Numerical Computation

The five model parameters for sodium have been evaluated with the help of eq. (5). The experimental elastic constants have been taken from the paper of Sharma & Joshi (1963) and the experimental zone boundary frequencies have been taken from the measurements of Wood *et al* (1962). The numerical values of the force constants are $A_1 = 1{\cdot}435$, $A_2 = 0{\cdot}883$, $K_1 = -0{\cdot}0541$, $K_2 = -0{\cdot}0194$,

$aK_e = -0{\cdot}3550$ all the units of 10^3 dyn cm^{-1}. The phonon dispersion curves are obtained from the solution of secular determinant (eq. (1)).

In the electrical resistivity calculations (eq. (6)) the integration over the solid angle Ω has been performed with the help of modified Houston's six direction procedure as elaborated by Betts *et al* (1956). The six directions used are (100), (110), (111), (210), (211) and (221). The distinction between the Normal and the Umklapp processes has been made by conservation law of the vectors while integrating over the scattering vector K. Earlier workers neglected the Umklapp contribution to the electrical resistivity (Bardeen 1937) but of late many workers (Bailyn 1960, Hasegawa 1964) have indicated that the electrical resistivity is dominated by the Umklapp processes and perhaps Normal processes can be neglected at low temperatures. The importance of the Umklapp process contribution to the low temperatures resistivity has been laid by Ekin (1971) and Ekin & Maxfield (1971). Bailyn (1960), Hasegawa (1964) and others have assumed that the Normal processes operate when the variable of integration U changes from 0 to 0·63 and the range 0·63 to 1 correspond to Umklapp processes. This separation between the two processes seems to be artificial and is valid only in the case of Debye model. In the present calculations the separation between the Normal and the Umklapp processes has been effected in a realistic manner by actually finding out which contributions are Umklapp and which are non-Umklapp (Kumar & Tolpadi 1975, Kumar 1975).

In the Normal process the electron wave vectors satisfy the relation $K = q$, where the phonon wave vector q, is made to lie in the first Brillouin zone. The elastic anisotropy has been accounted in the calculations by performing the integration over K numerically. The Umklapp processes occur subject to the selection rule $K = q + g$ where g is the reciprocal lattice wave vector. In this case K goes beyond the boundary of the first Brillouin zone but q is made to lie within it. The minimum value of K at which Umklapp process starts contributing to the total resistivity can be obtained from the geometry of the reciprocal lattice of a body centred cubic structure. The developed algorithm was simulated at IBM 360/44 computer system.

4. Results and Discussion

The computed phonon dispersion curves of sodium for selected values of the wave vectors in the symmetry directions are shown in fig. 1(a) along with the experimental data (Woods *et al* 1962). It is found that there is a good agreement between the theoretical and the experimental dispersion curves. In order to show a definite importance of G-function used in the present formulation, we have also calculated the dispersion curves for sodium using the electron-ion interaction from Sharma & Joshi (1963) model which uses the original Bardeen's G-function (Fig. 1(a)).

The calculations of the temperature variation of the electrical resistivity of Gugan 1960, 1962). It is seen that the computed and the experimental electrical

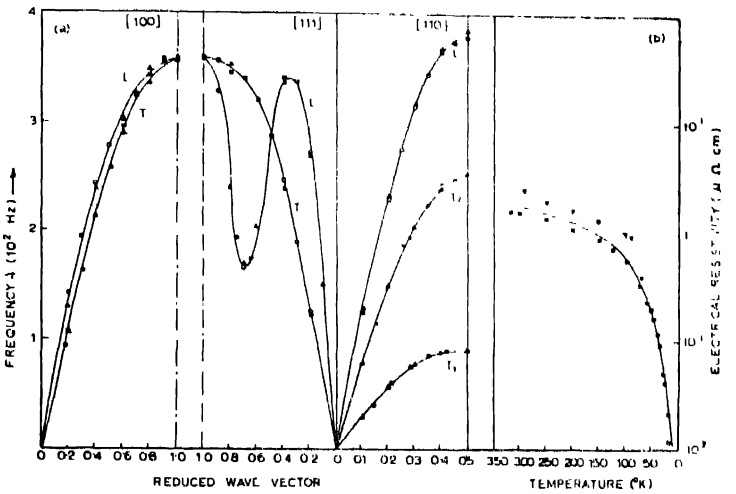

Fig. 1 (a) Phonon dispersion curves of sodium Full curves are present calculations. Δ are the calculations using original Bardeen's G-function O, , are experimental points (Wood *et al* 1962).

(b) Temperature variation of the electrical resistivity of sodium. Full curves are present calculation Δ are the calculations using original Bardeen's G function. [ ], O are the experimental points (Dugdale & Gugan 1962, 1960)

resistivity values iare in reasonable agreement. We have also plotted the computed electrical resistivity of sodium using the Bardeen's G-function in fig. 1(b). It is evident that the G-function used in the present algorithm has considerably improved the resistivity values.

The distinction between the Umklapp and non-Umklapp processes which has been made in a realistic manner in the present study has significantly affected the numerical values of the electrical resistivity, thereby emphasizing the fact that Umklapp contribution can not be neglected. A systematic increase in the Umklapp contribution to the total resistivity with the rise in temperature was observed. The Umklapp contribution in the case of sodium reaches a maximum value arround 15°K, its contribution to the total resistivity being roughly 45% at this temperature. Beyond 15°K, with the further increase in the temperature the Umklapp process contribution starts decreasing very slowly (being almost constant over a wide range of temperature) and at 300°K it drops to about 35% of the total electrical resistivity.

In addition to the influence of lattice vibrations on the resistivity there is an effect of the shape of the Fermi surface from sphericity enhances the proba-

bility of the Umklapp scattering of electrons, thereby causing an increase in the numerical values of the resistivity. This effect is expected to be significant in the case of other alkali metals but in the present study it does not concerns us since the Fermi surface of the sodium is effectively spherical (Shoenberg & Stile 1964). Moreover, our aim has been to see to what extent the temperature dependance of the electrical resistivity can be explained from phonon spectrum.

The discrepancies between theoretical and experimental electrical resistivity can be attributed to the neglect of the temperature variation of the elastic constants and other anharmonic effects. Sodium undergoes a martinistic type of phase transformation into a mixture of bcc and hcc structures at low temperature below about 37^{o}K and hence a rigorous comparison with the experimental values below its temperature is not feasible.

However, it is concluded that the proposed lattice dynamical model which incorporates a correct expression of Bardeen's G-function is able to give a satisfactory description of the experimental dispersion curves and provides a reasonable explanation of the temperature variation of the electrical resistivity.

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